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Life cycle assessment of different physical-chemical and biological technologies for biogas desulfurization in sewage treatment plants

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ABSTRACT

Biogas desulfurization can be carried out by physical-chemical or biological technologies. Biological technologies are generally described as more environmentally friendly than physical-chemical techniques, although it is difficult to find studies in which the associated environmental burdens are compared. Therefore, the aim of the work described here was to perform a life cycle assessment to compare the environmental impact of four common desulfurization technologies for hydrogen sulfide removal from biogas produced in sewage treatment plants. The desulfurization technologies studied were aerobic biotrickling filtration, anoxic biotrickling filtration, caustic chemical scrubbing and adsorption on impregnated activated carbon. The environmental assessment was extended with a Capital Expenditures (CAPEX) and Operational Expense (OPEX) analysis. Furthermore, an extra scenario (no desulfurization with biogas combustion) was compared with biogas desulfurization by aerobic biofiltration coupled to energy valorization (best-case treatment scenario). Results indicate that biological technologies are much more favorable in most environmental impact categories than physical-chemical technologies and, as expected, are more favorable than the non-treatment scenario. In all desulfurization technologies the most contributing stage to the total environmental burdens was the daily desulfurization operation. The chemical scrubber was the worst scenario except for the "Climate Change". In terms of the "Climate Change" (carbon dioxide equivalent emission), aerobic and anoxic desulfurization would emit 13.9 and 18.7 kg CO_2 kg^{-1} S-H_2S treated, respectively. The anoxic process could be improved to emit only 7.2 kg CO_2 kg^{-1} S-H_2S treated if the nitrate source was produced in a nitrification reactor built at the sewage treatment plant. Caustic chemical scrubber desulfurization and impregnated activated carbon desulfurization would emit 30.0 and 42.1 kg CO_2 kg^{-1} S-H_2S treated, respectively. To all impact categories, the contribution of sulfur compounds was less than 5%. The aerobic biotrickling filtration had the lower environmental impacts. However, the anoxic biotrickling filtration was the best approach if a nitrification reactor was already on-site.

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1. Introduction

Global Warming (GW) is currently a threat to human life and the environment. GW is caused by greenhouse gases such as methane (CH_4) and carbon dioxide (CO_2) (IPCC, 2014). In spite of its high contribution to GW, CH_4 can be used as a source of energy. For instance, one of the most important sources of CH_4 is biogas produced in anaerobic processes. Numerous substrates such as sewage sludge, livestock manure, waste from the agro-food industry and/or the organic fraction of municipal solid wastes can be used for biogas production (Deublein and Steinhauser, 2008; Ramírez et al., 2015). Biogas is an interesting energy source due to the high CH_4 concentration with low calorific values (LCV) ranging from 13.7 to 27.4 MJ Nm^{-3} for CH_4 concentrations ranging from 40 to 80%, respectively) (Ramírez et al., 2015). In particular, many Sewage

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Treatment Plants (STPs) produce biogas through the anaerobic digestion of sludge produced during wastewater treatment. Despite the fact that biogas purification and upgrading technologies are being developed for biogas use in the gas grid or as a vehicle fuel (Awe et al., 2017; Miltner et al., 2017), a large number of STPs use biogas for energy recovery in Combined Heat and Power (CHP) systems. For instance, the total electricity production from biogas in 2012 in the European Union was 46.4 TWh (64.7% from CHP plants and 35.3% from electricity-only plants) from more than 14,000 anaerobic digesters (EurObserv'ER, 2014). Nevertheless, the situation was different in Spain, with 88.3% of the total electricity production from biogas carried out in electricity-only plants (EurObserv'ER, 2014). In addition, it is common that biogas is flared in a number of situations, such as biogas overproduction or during CHPs malfunctioning or maintenance.

Biogas contains impurities such as hydrogen sulfide (H_2S) and ammonia (NH_3), two compounds that should be removed in order to avoid damage to CHP engines and to satisfy the European Directive on Industrial Emissions (Directive, 2010/75/EU). Biogas purification can be performed both physical-chemically and biologically. Biotrickling Filters (BTFs) for biogas biodesulfurization have proven to represent an effective technique to remove H_2S at pilot and full-scale under aerobic conditions (Rodriguez et al., 2014; Gabriel et al., 2013). However, under anoxic conditions only pilot and lab-scale studies have been carried out (Almenglo et al., 2016; Fernández et al., 2014; Brito et al., 2017). Physical-chemical processes such as chemical scrubbing using H_2O_2 (Krischan et al., 2012) and adsorption on impregnated activated carbon (Petersson and Wellinger, 2009) have commonly been carried out due to the existing knowledge, compactness and rapid startup of these technologies. However, biological processes appear to be more economical and environmentally friendly than physical-chemical processes (Gabriel and Deshusses, 2004; Tomàs et al., 2009). To the best of our knowledge, an environmental assessment by means of Life Cycle Assessment (LCA) has not been published to date on biogas desulfurization using BTFs. However, LCA has been applied to assess the impact of H_2S removal systems for odor treatment such as BTFs or biofilters (Alfonsín et al., 2013, 2015). For instance, Alfonsín et al. (2015) studied a range of odor removal technologies by LCA. A biofilter, a two-stage BTF, a chemical scrubber using NaOH-NaOCl, an activated carbon filter with granular activated carbon and a BTF coupled to an activated carbon tower were compared. Activated carbon was found to be the worst abatement technology based on climate change criteria due to the large demand of natural gas in the activated carbon production. Results obtained by Alfonsín et al. (2015) indicated that the use of an aerobic BTF would decrease carbon dioxide emissions by 84%. Moreover, Estrada et al. (2011) compared different odor abatement technologies in STPs using environmental and economic indicators. The odor abatement technologies were biofiltration, activated sludge diffusion, BTF, chemical scrubbing, activated carbon adsorption, regenerative incineration and a hybrid technology (BTF coupled with carbon adsorption). According to Estrada et al. (2011), biofiltration had the highest requirements in terms of materials (packing material) followed by chemical scrubbing because of the high empty bed residence time used in the biofilter and the high chemical reagents usage in the chemical scrubber.

In the present work, a comparative LCA study on four different desulfurization technologies was performed. Two technologies were biological processes: An aerobic BTF and an anoxic BTF, and other two were physical-chemical technologies: a chemical scrubber with hydrogen peroxide (H_2O_2) and an impregnated activated carbon adsorption unit. The four technologies were compared in order to select the most suitable technology from the point of view of environmental impact and operational and capital costs.

Anoxic desulfurization can be carried out using commercial nitrate (Almenglo et al., 2016; Brito et al., 2017) or nitrate produced in a nitrification bioreactor (Cano et al., 2016). Therefore, three different scenarios that can be found in a STP were selected, that is: (i) the construction of a nitrification bioreactor as a 'reference scenario', (ii) a nitrification bioreactor already built at the STP, (iii) chemical nitrate was used and then a nitrification bioreactor was not needed. Finally, the most suitable desulfurization technology combined with cogeneration was compared with a scenario in which there was no H_2S treatment and no energy recovery.

2. Materials and methods

2.1. System description

An anaerobic digester fed with sludge from a STP with a biogas flow rate production of 100 $m^3\ h^{-1}$ was considered for the analysis. The biogas composition was (% v v^{-1}): CH_4 (61.9), H_2S (0.3), CO_2 (37.5), N_2 (0.3) and NH_3 ($2.5 \cdot 10^{-2}$) (Deublein and Steinhauser, 2008). The process flow diagram for desulfurization technologies and the CHP system are show in Fig. 1.

Aerobic biological sulfide removal is a two-step process (Kuenen, 1975) that strongly depends on the amount of oxygen (O_2) supplied and transferred to the liquid phase. The first step leads to the production of elemental sulfur (S^0) (equation (1)) if oxygen supply is limiting. If enough O_2 is provided, the second step further proceeds to SO_4^{2-} production (equation (2)).

$$HS^- + 0.5O_2 \rightarrow S^0 + OH^- \quad (1)$$

$$HS^- + 2O_2 \rightarrow SO_4^{2-} + H^+ \quad (2)$$

Biological H_2S oxidation in anoxic BTFs works in a similar manner to aerobic BTFs. In this case, the electron acceptor is nitrate rather than O_2. Sulfide oxidation produces either S^0 (equation (3)) or SO_4^{2-} (equation (4)) depending on the amount of electron acceptor (nitrate) available in the system (Soreanu et al., 2008).

$$5H_2S + 2NO_3^- \rightarrow 5S^0 + N_2 + 4H_2O + 2OH^- \quad (3)$$

$$5H_2S + 8NO_3^- \rightarrow 5SO_4^{2-} + 4N_2 + 4H_2O + 2H^+ \quad (4)$$

Chemical scrubbing for sulfide removal using H_2O_2 as an oxidizing agent has been described by Couvert et al. (2008). Accordingly, the final oxidation product of sulfide is either S^0 (equation (5)), sulfite (SO_3^{2-}) (equation (6)) or SO_4^{2-} (equation (7)) depending on the H_2O_2 dose.

$$HS^- + H_2O_2 \rightarrow S^0 + H_2O + OH^- \quad (5)$$

$$S^0 + H_2O_2 \rightarrow SO_3^{2-} + H_2O + 2H^+ \quad (6)$$

$$SO_3^{2-} + H_2O_2 \rightarrow SO_4^{2-} + H_2O \quad (7)$$

Adsorption of H_2S on activated carbon is an efficient process for biogas desulfurization. However, impregnation of activated carbon with alkaline compounds such as sodium hydroxide (NaOH) is needed to improve H_2S removal. In this process both physical adsorption (equation (8)) and chemisorption (H_2S reaction with NaOH at carbon surface) (equations (9) and (10)) are involved (Yan et al., 2002; Sitthikhankaew et al., 2014).

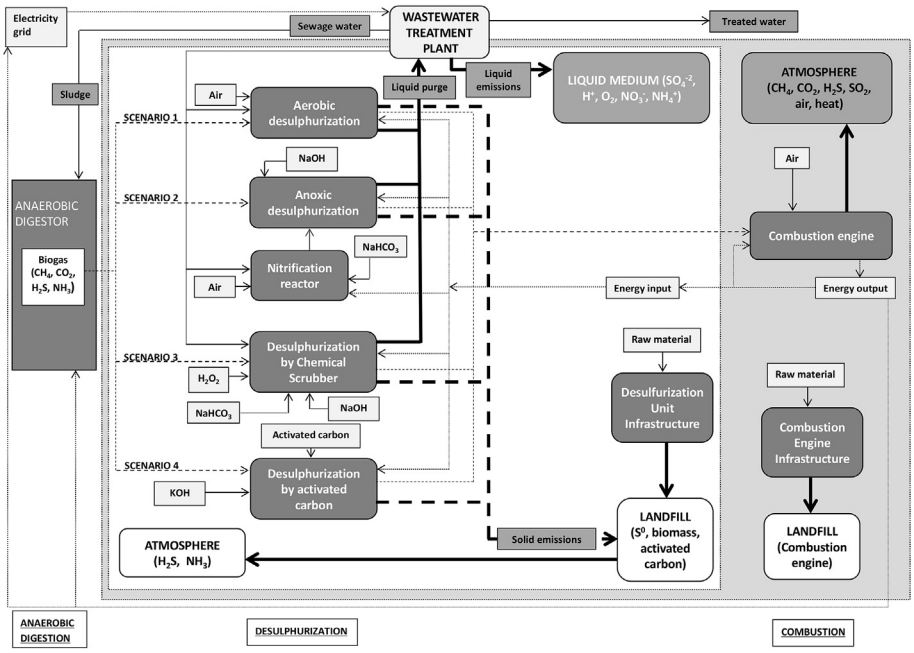

Fig. 1. Process flow diagram and boundaries considered for biogas desulfurization. Desulfurization boundaries were within the "white area" while combustion boundaries were within the "gray plus white area".

$$H_2S_{(g)} \rightarrow H_2S_{(ads)} \quad (8)$$

$$H_2S_{(g)} + NaOH_{(q)-C} \rightarrow NaHS_{(q)-C} + H_2O \quad (9)$$

$$H_2S_{(g)} + 2\,NaOH_{(q)-C} \rightarrow Na_2S_{(q)-C} + H_2O \quad (10)$$

where subscript "g", "ads" and "(q)-C" indicate that the compound is in the gas phase, physical adsorbed and impregnated or formed on the surface of the activated carbon, respectively.

2.2. Life-cycle assessment

2.2.1. Goal and scope

2.2.1.1. Goal of the study. The main goal of this study was to compare four different technologies for the removal of H_2S from biogas streams by means of LCA (Sc1.0, Sc2.0, Sc3.0, Sc4.0) (Table 1). Furthermore, three different alternatives for anoxic desulfurization were compared (Sc2.0, Sc2.1, Sc2.2). Finally, system expansion for the energy valorization of biogas by means of combustion in a CHP (with electricity recovery in Sc1.1, Sc2.3, Sc3.1, Sc4.1 and with electricity and heat recovery in Sc1.2) was performed and subsequently compared with direct biogas combustion in a flare without energy recovery (Sc5.0). The 12 LCAs are summarized in Table 1. It is worth mentioning that the audience of this study are biogas producers (mainly from STP) in addition to the scientific community.

2.2.1.2. Functional unit. The Functional Unit (FU) is a key parameter to perform the LCA since this describes the function of the system that is assessed. In the present case, we had an attributional LCA approach. Different FUs could be used to carry out the LCA, such as cubic meter of biogas treated, H_2S inlet concentration, H_2S inlet loading, kg of S-H_2S treated or kg of S-H_2S removed. Despite the fact that most LCA analyses for analogous processes use m^3 of biogas as FU (Alfonsín et al., 2013, 2015), such a FU does not take into account the H_2S inlet concentration or the removal efficiency. Therefore, kg of S-H_2S treated was considered as the FU in this study.

2.2.1.3. System boundaries

2.2.1.3.1. Desulfurization technologies. System boundaries for desulfurization processes (Sc1.0, Sc2.0, Sc3.0, Sc4.0) were defined within the white area in Fig. 1. Therefore, the desulfurization

Table 1
LCAs nomenclature.

Scenarios	Type
Sc1.0	Biogas desulfurization by aerobic biofiltration
Sc2.0	Biogas desulfurization by anoxic biofiltration considering the construction of a nitrification bioreactor
Sc2.1	Biogas desulfurization by anoxic biofiltration considering that the nitrification bioreactor is already built in the STP
Sc2.2	Biogas desulfurization by anoxic biofiltration using chemical nitrate
Sc3.0	Biogas desulfurization by chemical scrubbing
Sc4.0	Biogas desulfurization by impregnated activated carbon without regeneration
Sc5.0	Direct biogas combustion in a flare without energy recovery
Sc1.1	Scenario 1 expanded to electricity recovery
Sc1.2	Scenario 1 expanded to electricity and heat recovery
Sc2.3	Scenario 2 expanded to electricity recovery
Sc3.1	Scenario 3 expanded to electricity recovery
Sc4.1	Scenario 4 expanded to electricity recovery

process does not include the combustion engine and its related emissions. Purchased equipment cost (Table S1), chemicals, energy and natural resources supplied to operate the reactors were included within the limits. A cradle-to-grave LCA approach was considered for all scenarios.

2.2.1.3.2. Scenario analysis for anoxic desulfurization depending on the nitrate source. The first alternative for anoxic desulfurization, i.e., the reference scenario, was to build an extra bioreactor for nitrification (Sc2.0). The nitrification bioreactor was fed with reject water from the sludge dewatering systems of the STP (Carrera et al., 2011) to produce nitrate. Consequently, the nitrification process, the construction of the nitrification bioreactor and its disposal were considered in the LCA. Other alternatives could also be considered without building a nitrification bioreactor and these two subscenarios were: the nitrification bioreactor was available at the STP prior to implementing the anoxic BTF (Sc2.1) and the use of chemical nitrate from commercial suppliers (Sc2.2).

2.2.1.3.3. System expansion with biogas combustion. System boundaries for system expansion with biogas combustion were defined within the gray and white areas in Fig. 1. The system expansion with biogas combustion for electricity production was applied to the four different desulfurization technologies considered in the goal of this study, namely: Sc1.1, Sc2.3, Sc3.1 and Sc4.1 (Table 1). In addition, CHP for electricity recovery and heat production (Sc1.2) was included. It was considered that the energy recovered in the CHP substituted the energy provided by the Spanish electricity grid mix "Electricity mix, AC, consumption mix, at consumer, ES", data from Ecoinvent database 3.0 (Swiss Centre for LCI, 2013). The previous scenarios were compared with direct biogas combustion without desulfurization in a flare without energy recovery (Sc5.0).

2.2.1.4. Life cycle inventory (LCI) assumptions. The LCI included the following general assumptions:

- Solid waste related to the operation of all scenarios were dumped in landfill.
- Transport of solid waste to the landfill was not considered.
- Activated carbon is typically replaced rather than regenerated (Ryckebosch et al., 2011).
- There were three desulfurization processes that consume water (Sc1.0, Sc2.0 and Sc3.0) and these have environmental impacts due to the production of sulfate (SO_4^{2-}) in the liquid effluents. The rest of the pollutants present in the STP liquids were not modified. Therefore, the environmental impacts must be attributed to the STP operation and, consequently, were beyond the scope of the LCA.
- The elemental sulfur and the biomass accumulated in the packing material of the aerobic and anoxic BTF (Sc1.0 and Sc2.0) were disposed in a landfill site along with the exhausted activated carbon (NH_3 and H_2S adsorbed) from scenario Sc4.0.
- It was assumed that elemental sulfur, NH_3 and H_2S adsorbed by the activated carbon would not remain stable in the landfill site. On one hand, it was supposed that elemental sulfur would be reduced to H_2S and that a part of the H_2S and NH_3 from de active carbon would be desorbed and emitted to the atmosphere, the amounts of these emissions (H_2S and NH_3) were 40% of the total mass (Catalan Waste Agency, 2015; Spokas et al., 2006) while 60% were directed to the landfill biogas stream.
- All plants were designed by considering a lifespan of 20 years, which is the same as those of similar plants (Babbitt et al., 2009).

2.2.1.4.1. Aerobic desulfurization assumptions. The biogas stream is treated in an aerobic BTF (total volume of 10 m^3 and packed bed volume of 5 m^3) constructed with fiberglass reinforced plastic (FRP). Furthermore, auxiliary equipment such pumps, pipelines and energy to operate the system is required. Oxygen for biological H_2S oxidation was taken from atmospheric air by a jet-venturi. On using a jet-venturi the oxygen mass transfer can be enhanced by 26.7% compared to the conventional system using a blower (Rodriguez et al., 2014).

According to the biogas composition and flow rate considered in this study, a removal efficiency (RE) of 99.9% of the H_2S with an SO_4^{2-} production of 80% and concomitant S^0 production of 20% has been reported on operating with a dissolved oxygen load (DOL) per H_2S removed of 0.32 g O_2 g^{-1} S (López et al., 2016). The liquid that leaves the aerobic BTF is slightly acidic (pH 5.5). Consequently, NH_3 in the biogas stream is transferred from the gas phase to the liquid phase, with the formation of ammonium ions (NH_4^+) in the liquid emissions. All data attributed to the aerobic desulfurization are shown in Table 2.

2.2.1.4.2. Anoxic desulfurization assumptions. The biogas stream passes through an anoxic BTF (total volume of 10 m^3 and packed bed volume of 5 m^3) constructed with FRP. Similarly, H_2S RE of 99.9% with an SO_4^{2-} production of 69% and a concomitant S^0 production of 31% have been reported on operating at nitrogen sulfide (N:S) ratios of 1.4 mol N-NO_3^- mol^{-1} S-H_2S (Cano et al., 2015). Nitrate was produced in a nitrification bioreactor constructed with FRP with a volume of 8.3 m^3 (dilution rate of 0.029 h^{-1}). The size of the nitrification bioreactor was calculated considering an inlet ammonia concentration of 1 g N-NH_4^+ L^{-1} (Carrera et al., 2011), complete nitrification and an N:S ratio of 1.4 mol-N mol-S^{-1} in the anoxic BTF (Cano et al., 2015).

Furthermore, the same auxiliary equipment to that in Sc1.0 and energy is required. Sodium hydroxide (NaOH) is supplied to neutralize the pH in the anoxic BTF and sodium hydrogen carbonate ($NaHCO_3$) is supplied in the nitrification bioreactor as a carbon source. All data attributed to the anoxic desulfurization are shown in Table 2.

2.2.1.4.3. Chemical scrubbing desulfurization assumptions. The biogas stream is treated in a chemical scrubber (total volume of 1.69 m^3 and packed bed of 0.84 m^3) constructed with FRP. In this study, an H_2S RE of 99.9% for complete conversion of H_2S to SO_4^{2-} is feasible on operating with excess H_2O_2 (Krischan et al., 2012).

In addition to the auxiliary equipment of Sc3.0 and energy to operate, H_2O_2, $NaHCO_3$ and NaOH is supplied for H_2S absorption and oxidation. All data attributed to the chemical scrubbing desulfurization are shown in Table 2.

2.2.1.4.4. Assumptions for impregnated activated carbon desulfurization without regeneration. The biogas stream passes through an activated carbon desulfurization unit with a packed bed volume of 6.0 m^3 constructed with FRP. The activated carbon is impregnated with potassium hydroxide (KOH) at 5 wt% to improve H_2S removal up to 99.9% (Wu et al., 2005; Kawano et al., 2008). All data attributed to the activated carbon and impregnation process with KOH at high temperature (800 °C) are shown in Table 2.

2.2.1.4.5. Assumptions for system expansion. In Sc5.0, heat waste and gases such as SO_2, nitrogen oxides (NO_x) and CO_2 is produced and emitted to the atmosphere. All data attributed to the direct biogas combustion, its infrastructure and its disposal are shown in Table 2.

In the scenarios with electricity recovery (Sc1.1, Sc2.3, Sc3.1, Sc4.1) and electricity and heat recovery (Sc1.2), the desulfurized biogas stream is coupled to a combustion engine (400 kW, model TCG, 2016 V08 C) operating at 50 Hz and constructed with 5.3 tons of steel (MWM, 2017). In this case, the total recoverable energy considering the combustion engine depends on the type of engine: 27% for electrical energy recovery and 66% for CHP recovery (Kang et al., 2012). The temperature of the exhausted gases is 1280 °C for electrical energy recovery and 1190 °C for electricity and heat

Table 2
Life Cycle Inventory for the system expand.

	Units (UF^{-1})	Scenario								
		Sc1.0	Sc1.1	Sc2.0	Sc2.3	Sc3.0	SC3.1	Sc4.0	Sc4.1	Sc5.0
Inputs										
Material										
Construction										
Steel	g	3.8	80.7	3.8	80.7	3.8	80.7	3.8	80.7	7.2
Steel Pipe	g	0.7	1.5	0.7	1.5	0.7	1.5	0.7	1.5	0.7
FRP	g	8.2	8.2	12.3	12.3	0.3	0.3	1.7	1.7	–
PVC	g	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	–
Polypropylene	g	1.2	1.2	1.2	1.2	0.6	0.6	0.6	0.6	–
Stirring system	m^3	–	–	$9.6\cdot 10^{-5}$	$9.6\cdot 10^{-5}$	–	–	–	–	–
Packing Material										
Polypropylene Pall Ring	m^3	$1.2\cdot 10^{-4}$	$1.2\cdot 10^{-4}$	$1.2\cdot 10^{-4}$	$1.2\cdot 10^{-4}$	$1.1\cdot 10^{-5}$	$1.1\cdot 10^{-5}$	–	–	–
Activated carbon										
Non-regenerative	kg	–	–	–	–	–	–	4.1	4.1	–
Daily operations										
Chemicals										
NaOH	moles	–	–	3.0	3.0	187	187	–	–	–
$NaHCO_3$	moles	–	–	$2\cdot 10^{-2}$	$2\cdot 10^{-2}$	13,411	13,411	–	–	–
H_2O_2	kg	–	–	–	–	9.9	9.9	–	–	–
KOH	g	–	–	–	–	–	–	204	204	–
Water										
Sewage water	m^3	12.2	12.2	0.6	0.6	11.2	11.2	–	–	–
Energy										
Electricity	kWh	20.4	209	27.4	216	10.2	198	0.1	188	–
Heat	kWh	–	–	–	–	–	–	0.8	0.8	–
Natural Sources										
Air	m^3	26.5	1752	27.2	1799	–	1725	–	1725	1725
Outputs										
Daily operations										
Gaseous emissions										
H_2S	mg	84.6	95.2	128	132	–	319	424,722	424,851	10,629
CH_4	kg	–	1.0	–	1.0	–	1.0	–	1.0	1.0
NH_3	g	–	–	–	–	–	–	17.7	17.7	0.4
NO_x	g	–	–	–	–	–	–	–	1.6	118
SO_2	g	–	2.0	–	2.0	–	59.3	–	29.1	1978
Liquid emissions										
O_2	g	97.7	97.7	–	–	89.3	89.3	–	–	–
SO_4^{2-}	g	2397	2397	2071	2071	2906	2906	–	–	–
NH_4^+	g	46.8	46.8	46.8	46.8	46.8	46.8	–	–	–
NO_3^-	mg	–	–	17.0	17.0	–	–	–	–	–
H^+	mg	38.9	38.9	–	–	–	–	–	–	–
Energy										
Electricity	kWh	–	444	–	444	–	444	–	444	–
Energy waste										
Heat waste	kWh	–	666	–	666	–	666	–	666	1035
Waste dumped										
Biomass	g	52.6	52.6	79.7	79.7	–	–	–	–	–
Steel	g	4.5	82.2	4.5	82.2	4.5	82.2	4.5	82.2	7.9
FRP	g	8.2	8.2	12.3	12.3	0.3	0.3	1.7	1.7	–
PVC	g	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	–
Polypropylene	g	1.2	1.2	1.2	1.2	1.2	1.2	0.6	0.6	–
Activated Carbon	kg	–	–	–	–	–	–	4.1	4.1	–
Stirring system	m^3	–	–	$9.6\cdot 10^{-5}$	$9.6\cdot 10^{-5}$	–	–	–	–	–

recovery (Kang et al., 2012). A 15% oxygen excess in air for biogas combustion is needed for 99% CH_4 combustion. In addition, electricity, heat waste and gases such as SO_2, nitrogen oxides (NO_x) and CO_2 are produced and emitted to the atmosphere. Finally, the combustion engine is sent to landfill. All data attributed to the biogas combustion for electricity production, its infrastructure and disposal are shown in Table 2.

2.2.1.5. Data collection and data quality. The basis for inputs and outputs to carry out the LCA are provided in Table 2. Data collection is one of the most important steps to carry out a LCA successfully. The research groups involved in this study have wide experience at pilot scale in aerobic and anoxic desulfurization, and experimentally obtained the data used in this study in previous works (Tomàs et al., 2009; Rodriguez et al., 2014; Cano et al., 2015; Almenglo et al., 2016; López et al., 2016). Chemical scrubbing and impregnated activated carbon are well-known technologies; therefore, data were collected from peer-reviewed publications. Specifically, data for chemical scrubbing desulfurization were obtained from Couvert et al. (2008) (lab-scale) and Krischan et al. (2012) (pilot-scale), and data for desulfurization using impregnated activated carbon were obtained from Wu et al. (2005) and Kawano et al. (2008), who described impregnation methods with KOH. Data for biogas combustion were obtained from a full off-design analysis of a CHP system carried out by Kang et al. (2012). Data required for the purchased equipment cost (Table S1) of the desulfurization units and data concerning auxiliary equipment, were obtained from pilot- and full-scale units used by the two groups involved in this study (Gabriel and Deshusses, 2004; Tomàs et al., 2009; Rodriguez et al., 2014; Almenglo et al., 2016). Data required for the

combustion units (flare and CHP engine) were obtained from Tomás et al. (2009) and from the company MWM GmbH (MWM, 2017). To complete the LCI, the Ecoinvent database 3.0 (Swiss Centre for LCI, 2013) was used.

2.2.2. Life-cycle inventory

Only mandatory steps defined by the UNE-EN ISO 14040–14044 regulation for impact assessment (International Organization for Standarization, 2006a, b), namely classification and characterization. Values of the LCI used to perform the LCAs are shown in Table 2 for (i) only biogas desulfurization (Sc1.0, Sc2.0, Sc3.0, Sc4.0) and (ii) the system expansion with electricity recovery (Sc1.1, Sc2.3, Sc3.1, Sc4.1) and the direct combustion of biogas in a biogas flare without desulfurization (Sc5.0).

The energy recovery by combustion involves a preliminary desulfurization step to avoid further problems in the combustion engine (Sevimoğlu and Tansel, 2013). Therefore, environmental impacts attributable to desulfurization must also be attributable to combustion with electricity recovery.

2.2.3. Life-cycle impact assessment

SimaPro 8.0.4.30 software was used in this study. The impact assessment method used was Recipe Midpoint (H). This method was used because it harmonizes two usual impact assessment methods such as CML and Eco-indicator 99 in one (Goedkoop et al., 2008). In order to compare the different scenarios, the six categories with the highest impact values were selected for the biological scenarios. Moreover, it was considered the "Climate Change" category was considered to calculate the carbon dioxide equivalent emission of each desulfurization technology and its improvement installing a CHP for energy recovery.

2.3. CAPEX and OPEX analysis

Economic factors are often prioritized over environmental factors. Accordingly, environmentally friendly processes should be economically viable as well as sustainable (Singh et al., 2013). Therefore, analysis of capital costs and operating costs attributed to desulfurization processes are a key factor to verify the possibility of its further implementation. Thus, each desulfurization process LCA was complemented with a CAPEX and OPEX analysis.

The economic model was carried out in two steps. First, CAPEX of the different alternatives and desulfurization scenarios were assessed. Then, OPEX were calculated to complete the economic assessment by analyzing the maintenance and operating costs of the desulfurization processes.

The percentages of Fixed-Capital Investment (FCI) for direct and indirect costs was based on the range published by Peter and Timmerhaus (1991). Direct FCI costs included purchased equipment (30%), installation (14%), instrumentation and control (installed) (8%), piping (installed) (6%) and electrical systems (installed) (8%) costs. Buildings, yard improvements and land were supposed negligible because the STP was previously built. Indirect FCI costs included engineering and supervision (13%), construction expenses (8%), contractor's fees (3%) and contingency (10%) costs. Annual maintenance costs were considered as 1.5% of the total direct cost (Aguilera and Gutiérrez-Ortiz, 2016).

The Purchase Equipment Costs (PEC) were provided by Bioconservacion S.A. It was needed to resize the cost using equation (11).

$$C_S = C_0 \cdot \left(\frac{X_S}{X_0}\right)^n \quad (11)$$

where X_s was the actual scale, X_0 was the initial scale, C_s was the purchase cost of the equipment with the actual cost, C_0 was the purchase cost of the equipment with the initial scale and n is the scaling factor. For the present study, it was supposed a value of 0.67 (Starr et al., 2015).

Moreover, all costs were based on July 2017 prices. Chemical Engineering Plant Cost Index (CEPCI) was used as index cost to update capital costs (Peter and Timmerhaus, 1991). Materials (chemical, packing, activated carbon, etc.), utilities cost (electricity and heat) and labor cost were updated using the general Consumer Price Index (CPI) (INE, Spanish Statistical Office, 2017, Spain).

3. Results and discussion

3.1. Environmental assessment of desulfurization technologies

The result obtained in each impact category per FU was the sum of the construction of the desulfurization unit (CDU) plus the daily desulfurization operations (DDO) plus the final disposal of the desulfurization unit and its auxiliary equipment in a landfill (FDD). The quantification data for all impact categories are shown in Fig. 2.

For the two conventional physical-chemical processes, the contribution attributable to CDU and FDD was almost negligible. However, for the two biological scenarios, the contribution attributable to CDU and FDD was measurable but it was not as large as for DDO.

In the cases of Sc1.0 and Sc2.0, several impact categories such as

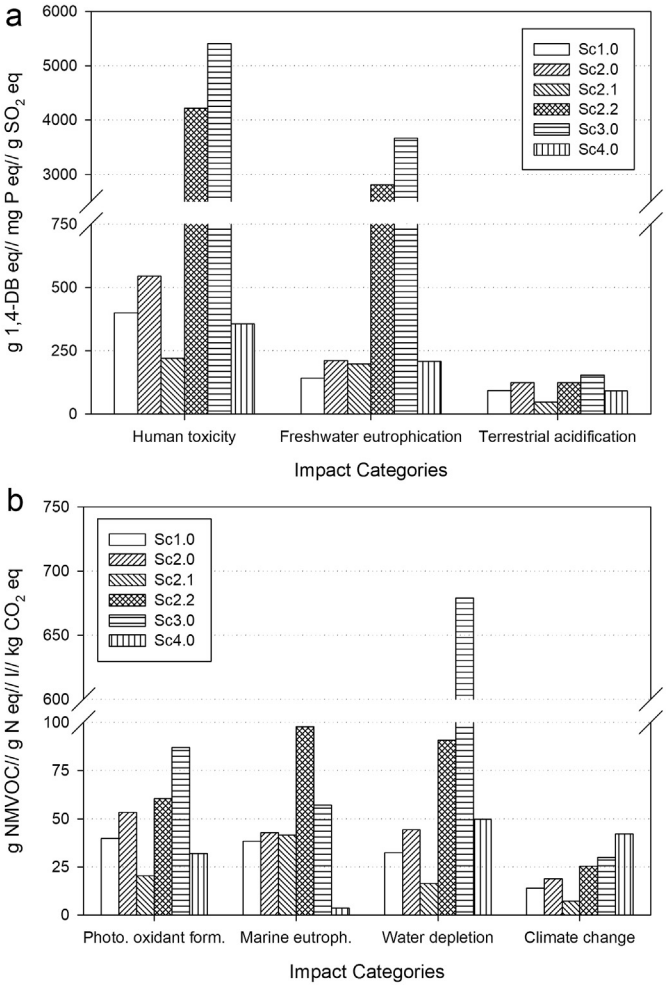

Fig. 2. Impact categories for desulfurization scenarios.

"Climate Change", "Terrestrial Acidification", "Marine Eutrophication", "Photochemical Oxidation Formation" and "Human Toxicity" were generated mainly because of the resource use and emissions involved in DDO. All these categories except for "Marine Eutrophication" and "Photochemical Oxidation Formation" were above all caused by electricity consumption since more than 90% of the impacts were due to their use.

Otherwise, "Marine Eutrophication" was caused by the nitrogen emissions in the liquid phase. Other environmental effects were generated due to the combination of DDO, principally, plus CDU, such as "Freshwater Eutrophication". In relation to "Freshwater Eutrophication", effects of this category were mainly provoked (80%) by the biomass disposal in the landfill (DDO). Related to CDU, plastics requirements for the construction of infrastructures and processes are inducers, at least, of 10% of the total impact impacts in this category. It was noteworthy that no significant impact was appreciated for "Freshwater Eutrophication" because of the low nitrate emissions in the liquid phase (DDO) (Table 2).

"Water Depletion" of 32.4 and 44.4 L for both scenarios Sc1.0 and Sc2.0, respectively, had a negative impact on the environment because of the electricity consumption in DDO. In this impact category, the CDU had a negative value because of the molding process of the FRP, −0.8 L and −0.3 L for Sc1.0 and Sc2.0, respectively. The value of FDD was negligible.

Most impact categories for Sc3.0 and Sc4.0 were generated due to DDO.

The highest emissions occurred in Sc3.0 except for the "Climate Change", which was slightly higher in Sc4.0 (Fig. 2b). This scenario (Sc3.0) was considered to be the worst desulfurization alternative. The main inducer corresponded to H_2O_2 production, which created more than 80% of the total impact in the following categories: "Freshwater Eutrophication", "Marine Eutrophication", "Human Toxicity" and "Water Depletion". Then the combination of H_2O_2 and NaOH production created more than 70% of the total impact in the following categories: "Climate Change" (42% H_2O_2 production and 35% NaOH production), "Terrestrial Acidification" (38.7% H_2O_2 production and 32% NaOH production) and "Photochemical Oxidation Formation" (47% H_2O_2 production and 31% NaOH production).

Sc1.0 and Sc2.0 were quite similar (Fig. 2b), but Sc2.0 had a higher impact in all the impact categories due the construction of the nitrification bioreactor. Both scenarios (Sc1.0 and Sc2.0) had a lower impact than Sc4.0 in all of the categories except for "Terrestrial Acidification", "Marine Eutrophication", "Photochemical Oxidation Formation" and "Human Toxicity". The main inducer in Sc4.0 was the carbon production and impregnation process with KOH.

The "Climate Change" was considered to be the most important category since the goal of the desulfurization process was to facilitate the biogas energy recovery to avoid burning other fuels that would increase the amount of CO_2 in the environment. On comparing the desulfurization technologies, the most suitable scenario was Sc1.0 because the amount of CO_2 equivalents emitted would be 13.9 kg for the FU employed. This emission would increase for Sc2.0 up to 18.7 kg and up to 30.0 kg for Sc3.0. Finally, the worst situation corresponded to Sc4.0 because of its high CO_2 equivalent emission, 42.1 kg. Concerning the origin of the CO_2 equivalent emissions produced because of the desulfurization processes, the DDO was found to be the main inducer for all scenarios. In Sc1.0 and Sc2.0 electricity consumption would produce more than 98% of the emissions. In Sc2.0 the main inducer would be the electricity consumption in the nitrification bioreactor because CO_2 equivalent emissions made up 61% of the total CO_2 equivalent emissions. In Sc3.0 H_2O_2 production corresponded to more than 42% of the total CO_2 equivalent emissions produced by chemical scrubbing with H_2O_2. Large amounts of other chemicals such as $NaHCO_3$ and NaOH would also be required to carry out the desulfurization, as shown in Table 2. These chemicals were the second highest inducer of CO_2 equivalent emissions because they corresponded to more than 35% of the total CO_2 emissions. Finally, it is also worth highlighting the importance of electricity consumption in the desulfurization by chemical scrubbing with H_2O_2, which corresponded to more than 22% of the total CO_2 emissions. In Sc4.0 the production and preparation of activated carbon were the cause of the CO_2 equivalent emissions. The sum of these flows represented 26% of the total CO_2 equivalent emissions. The alkaline impregnation (4% of the total impacts) was also significant. The remaining 70% were the result of a large number of flows with low CO_2 equivalent emissions (less than 1% of the total for each one). Moreover, carbon production and preparation mainly affected the "Human Toxicity" and "Marine Eutrophication". The main "Human Toxicity" inducer in Sc4.0 was the use of KOH to activate the carbon, which caused more than 50% of the total "Human Toxicity". In addition, the production of carbon from underground and pit mining and the electricity consumption were also very significant and caused more than 44% of the total impact. Moreover, the sourcing of carbon from underground and pit mining led to emissions that corresponded to around 14% out of the total "Marine Eutrophication", while activation with KOH caused 24% of the total emissions.

Results obtained by Alfonsín et al. (2015) indicate that the use of an aerobic BTF could decrease the emissions of carbon dioxide by up to 84% if an aerobic BTF was used instead of activated carbon. In the present study, the decrease was also considerable, around 67%, and this lower value was due to higher amounts of energy and materials needed for H_2S removal from biogas instead of air.

In Sc1.0 the "Marine Eutrophication" was mainly due to NH_4^+ emissions, which are responsible for more than 90% of the total impact. The "Human Toxicity" was mainly due to electricity consumption, which caused more than 95% of the total emissions.

In Sc2.0 the "Marine Eutrophication" and "Human Toxicity" increased up to 42.7 g N eq and 546 g 1-4-DB eq, respectively. As in Sc1.0, NH_4^+ emissions were an important generator in the "Marine Eutrophication". Additionally, there would be nitrate emissions that would create a less favorable value than in Sc1.0. As in Sc1.0, the "Human Toxicity" was mainly due to electricity consumption, which caused more than 93% of the total emissions.

Estrada et al. (2011) also studied the environmental impact of a desulfurization process for odor abatement. According to Estrada et al. (2011), chemical scrubbing has the highest requirements in terms of reagents, which is consistent with the results of the current study.

For all scenarios, a low contribution (lower than 5%) due to the presence of sulfur compounds (S^0, SO_4^{2-} and H_2S) was found.

3.1.1. Scenario analysis for anoxic desulfurization depending on the nitrate source

In Sc2.1 the CO_2 the emissions were reduced to 7.2 kg CO_2 equivalents per FU. In this respect, a 61.4% decrease compared to Sc2.0 was produced in the impact of "Climate Change" because a nitrification bioreactor was not implemented due to the presence of nitrate in the water from the STP. In addition, other impact categories such as "Terrestrial Acidification", "Freshwater Eutrophication", "Marine Eutrophication", "Human Toxicity", "Water Depletion" and "Photochemical Oxidant Formation" decreased and these decreases ranged from 3% for "Marine Eutrophication" to 63% for "Water Depletion". Once again, such decreases were due to the absence of the nitrification bioreactor in Sc2.1. The impact distribution corresponding to Sc2.0 (column bar lower than 100% signifies an impact reduction) is represented in Fig. 3. Therefore,

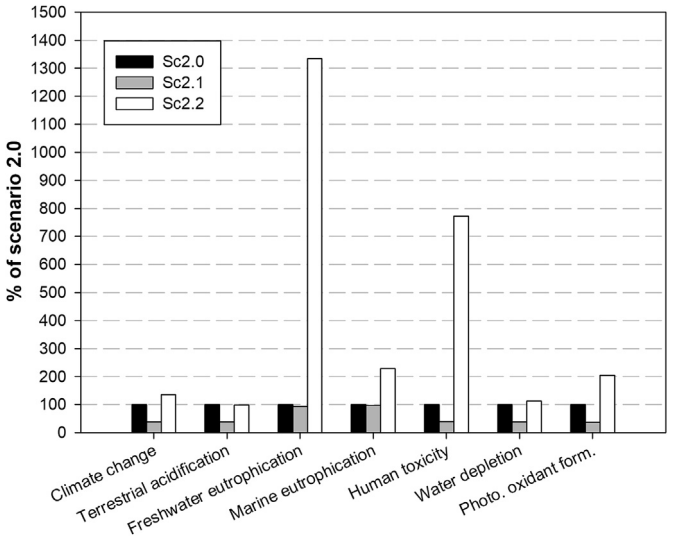

Fig. 3. Comparison of the three different scenarios for anoxic desulfurization.

anoxic desulfurization was a good alternative to aerobic desulfurization, as can be seen from Figs. 2 and 3.

In Sc2.2 the CO_2 emissions increased up to 25.3 kg CO_2 equivalents. Therefore, an increase of 35% in the impact of "Climate Change" was produced. In addition, other impact categories such as the "Water Depletion" and "Photochemical Oxidant Formation" increased up to 105% and 14%, respectively. These small increments were mainly due to the chemical nitrate usage since more than 70% of the total impact in the categories discussed above were associated with chemical nitrate usage. Despite these small increments, other categories such as "Freshwater Eutrophication", "Marine Eutrophication" and "Human Toxicity" increased in the range from 129% in the case of "Marine Eutrophication Category" to 1234% for "Freshwater Eutrophication". Therefore, the use of commercial nitrate was not a suitable alternative to Sc2.0, as shown in Fig. 3. In Sc2.2, the environmental behavior of the anoxic desulfurization would resemble a physical-chemical process (Sc3.0), as can be seen from the results in Fig. 2.

3.2. Environmental assessment of system expansion with biogas combustion

Four different LCAs for biogas desulfurization and biogas combustion in a CHP with electricity recovery were performed (Sc1.1, Sc2.3, Sc3.1, Sc4.1) and one LCA was considered for the direct biogas combustion in a flare without energy recovery (Sc5.0).

Results obtained showed an analogous behavior to the desulfurization study without system expansion (Table S2). Therefore, the scenario of choice was Sc1.1, which was quite similar to Sc2.2, and the worst scenario was Sc3.1. The results for Sc1.1 and Sc5.0 are shown in Table 3.

Table 3
Impact category results for the two alternatives for biogas energy: electrical (Sc1.1) and heat and electrical (Sc1.2) and for direct biogas combustion in a flare without energy recovery (Sc5.0).

Impact category	Units (FU^{-1})	Sc1.1	Sc1.2	Sc5.0
Climate Change	kg CO_2 eq	−230	−411	26.0
Terrestrial Acidification	g SO_2	−1729	−2323	2046
Freshwater Eutrophication	mg P eq	676	−6862	54.8
Marine Eutrophication	g N eq	15.5	5.5	4.8
Human Toxicity	g 1,4-DB eq	−6410	−16,106	44.8
Water Depletion	L	−644	−830	0.3
Photo. oxidant form.	G NMVOC	−726	−1049	290

A negative value for the impact category signifies a positive impact on the environment. Therefore, in Sc1.1 five impact categories had a positive impact on the environment (Table 3). This positive impact on the environment is mainly due to the electricity production by the CHP (Wang et al., 2014), which allows reducing the electricity net consumption (avoided product).

In Sc5.0 all impact categories had a positive value and therefore a negative impact on the environment because of the zero energy recovery and due to the presence of SO_x. Fig. 4 shows that the main inducer of the negative impacts is the biogas combustion process ("Climate Change", "Terrestrial Acidification", "Marine Eutrophication" and "Photochemical Oxidant Formation") and the construction of the combustion unit ("Freshwater Eutrophication", "Human Toxicity", "Freshwater Eutrophication" and "Water Depletion").

Fig. 4 shows the contribution of the three main stages for the

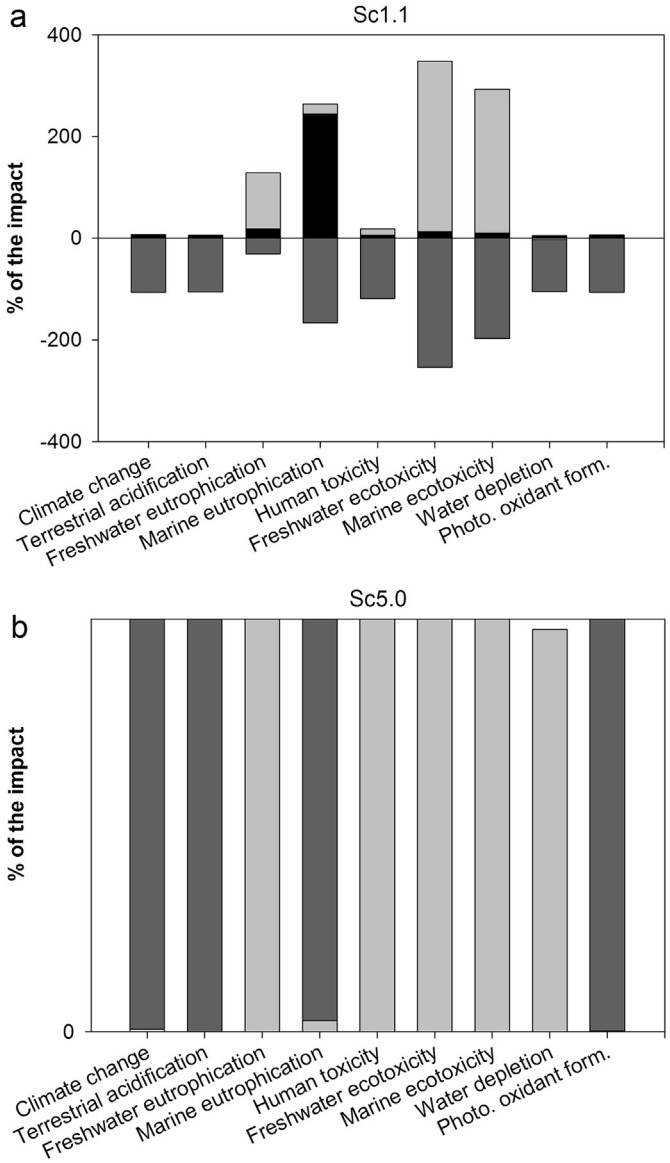

Fig. 4. Results of the system expansion study for energy recovery. Black: DDO, Light Gray: CCU, Dark Gray: DCO. Fig. 4 a: aerobic desulfurization plus combustion with energy recovery (Sc1.1); Fig. 4 b: Direct biogas combustion in a flare without energy recovery (Sc5.0).

Sc1.1 and Sc5.0 for each impact category. The 3 stages selected were DDO, "the construction of the combustion unit" (CCU) and "the daily combustion operations" (DCO). These stages were selected because they were the main inducer stages.

Sc5.0 was very harmful for the environment (Fig. 4b and Table 3). In the case of the "Climate Change", the emissions of CO_2 equivalents per FU were 26 kg. Nevertheless, a previous desulfurization unit would save up to 230 kg CO_2 per FU in Sc1.1. The main inducer of the negative impacts for the combustion of biogas directly in a flare was the gaseous emissions (99% of the total). On the other hand, the main inducer of the positive impacts for the rest of scenarios was electricity recovery, which would be able to balance the energy consumption to combust the biogas. It is also important to highlight the results obtained in the "Terrestrial Acidification". It would be possible to save up to 1729 g SO_2 per FU in Sc1.1 compared with the emission of 2046 g SO_2 per FU in Sc5.0. For the "Terrestrial Acidification" the main inducer of the negative impacts were the gaseous emissions (99% of the total), as in the "Climate Change". The biogas desulfurization plus electricity recovery in a CHP (Table S2) was able to balance the negative impacts of the emission of SO_2 in the CHP due to H_2S combustion and the energy consumption to combust the biogas. Related to scenarios Sc1.1 and Sc2.3 (Table S2), if the nitrification bioreactor is already built at the STP both BTFs (anoxic and aerobic) in conjunction with electricity recovery had similar environmental impacts.

Moreover, electrical energy recovery was compared with electrical and heat energy recovery in a CHP. For biogas energy recovery, only electricity production was considered because the majority of the plants located in Spain that recover energy produce electricity rather than electricity and heat. However, most plants in the rest of Europe recover energy as electricity and heat (EurObserv'ER, 2014). Thus, a sensitivity analysis was performed to compare electrical energy recovery in an engine that has a recuperative cycle gas turbine producing heat and electrical energy (Kang et al., 2012).

A comparison of the impacts on the environment for combustion in an engine with electrical energy recovery (Sc1.1), for combustion in a CHP with heat and electrical energy recovery (Sc1.2) and for direct biogas combustion in a flare without energy recovery (Sc5.0) can be made from the results in Table 3. Aerobic biodesulfurization was selected because it was previously identified as the most favorable technology to desulfurize biogas from an environmental perspective. Aerobic desulfurization and its infrastructure and disposal for both types of energy recovery are considered in Table 3. The heat and electricity recovery (Sc1.2) was better in all impact categories than for the recovery of electricity only (Sc1.1). However, not all categories improved in the same way. A comparison between Sc1.1 and Sc1.2 showed an improvement for the "Climate Change" up to 78% because of the heat recovery. Moreover, the "Water Depletion", "Terrestrial Acidification", "Photochemical Oxidant Formation" and "Marine Eutrophication" were improved by 29, 34, 44 and 64%, respectively. Additionally, some impact categories were modified from a negative impact (Sc1.1) to a positive impact on the environment (Sc1.2), such as "Freshwater Eutrophication". Finally, the "Marine Eutrophication" improved despite it still being a negative impact on the environment.

3.3. CAPEX analysis of biogas desulfurization

Table 4 shows the PEC associated with the desulfurization plants updated to July 2017 according to its scale and CEPCI. The costs of the columns used in the BTFs, chemical scrubber and activated carbon column was obtained using equation (11) from initial scale of 1.5 m^3 vessel of FRP (4500 €, January 2016) (Bioconservacion S.A, 2016). The actual scale of each desulfurization technology was: BTFs 10 m^3, chemical scrubber 1.7 m^3 and activated carbon column 6 m^3. The other PECs were obtained from suppliers for the necessary scale (Table 4, Table S1).

The highest total purchase cost was for Sc2.0 due to the installation of the nitrification bioreactor and its auxiliary equipment (air diffuser and blower) with an extra purchase equipment cost of 18,665 € (Table S1). In fact, if the nitrification bioreactor is already built in the STP the purchase equipment of Sc1.0, Sc4.0 and Sc2.1/Sc2.2 are quite similar, between 28,882 and 35,745 € (Table 5). The lowest PEC was for Sc3.0 due to its lower column size (21,949 €). FCI costs were between 73,164 and 174,932 €. Similar FCI were obtained in a study for biogas desulfurization (190 $m^3\ h^{-1}$, inlet H_2S concentration of 3000 ppmv) using a BTF (175,000 €) and activated carbon (105,000 €) (de Arespacochaga et al., 2014).

3.4. OPEX analysis of biogas desulfurization

Table 6 shows the utility costs for the desulfurization plants operation. Costs were updated to July 2017 according to the CPI in Spain. Additionally, a 25% of the total salary for a technician employee (9317.46 €) was considered based on current workforce price in Spain (INE, 2017).

All costs are presented per 1 kg of S-H_2S treated (FU used in the LCA) and per m^3 of biogas treated. Table 7 shows that operation of biological processes (Sc1.0, Sc2.0, Sc2.1 and Sc2.2) is much more economical than physical-chemical processes. The use of a physical-chemical desulfurization unit could increase costs by up to 16 times if a chemical scrubber was used instead of an aerobic BTF or by 13 times if a chemical scrubber was used instead of an anoxic BTF (Sc2.0).

Comparison of aerobic (Sc1.0) and anoxic desulfurization processes (Sc2.0) shows that the difference is the cost related to the electron acceptor consumption. While aerobic desulfurization uses O_2 from air, anoxic desulfurization uses nitrate supplied from a nitrification bioreactor. The implementation of a nitrification bioreactor implies an operational cost increase of 0.43 c€ per m^3 of biogas treated (energy consumption) and such an increase can be assumed if dilution of biogas with air is not required. In contrast, if the nitrification reactor is already built at the STP (Sc2.1) the operational cost is the lowest (1.71 c€ per m^3 of biogas treated) and the use of chemical nitrate (Sc2.2) had a similar cost to the implementation of the nitrification bioreactor (2.48 c€ per m^3 of biogas treated). Despite of the similar operational cost of Sc2.0 and Sc2.2, the environmental impacts of Sc2.2 was higher than Sc2.0 (Fig. 2). Therefore, Sc2.0 is more advantageous from a sustainable point of view.

Tomàs et al. (2009) carried out an economic study of biogas desulfurization using an aerobic BTF treating 3000 ppmv of H_2S for a biogas flow rate of 80 $m^3\ h^{-1}$. The cost per m^3 of biogas was 1.43 c€ (July 2017, data updated using the CPI), which is similar to the value obtained in this study (2.04 c€ per m^3 of biogas treated). Analogously, Fernández et al. (2014) performed an economic study on an anoxic BTF using chemical nitrate supplied from $NaNO_3$. The cost per m^3 of biogas increased up to 1.60 c€ (July 2017, data updated using the CPI), i.e., 12% more than the cost reported by Tomàs et al. (2009) and 0.88 c€ less than the value obtained in this study using commercial nitrate (Sc2.2).

Aguilera and Gutiérrez-Ortiz (2016) studied the operational cost of desulfurizing biogas by different processes. For activated "carbon siloxa", treating 447 ppmv of H_2S in a biogas stream of 500 $m^3\ h^{-1}$, the cost per kg of S-H_2S treated was 13.9 € (July 2017, data updated using the CPI) – a value close to that obtained in this study (13.65 € per kg of S-H_2S treated).

The operational cost was mainly due to the energy consumption and workforce (>85%) in Sc1.0, Sc2.0 and Sc2.1 (Table S3), while in

Table 4
Purchase equipment costs associated with the desulfurization.

Equipment[a]				
Description	Cost (€ Units^{-1})	CEPCI[a]	Costs (July 2017) (€ Units^{-1})	Source
BTF made with FRP (total volume 10 m^3, packing bed volume 5.3 m^3, diameter 0.9 m).	15,940 (Jan. 2016)	640.5	17,090	Bioconservacion S.A. (2016)
Scrubber made with FRP (total volume 1.7 m^3, packing bed volume 1.1 m^3, diameter 0.7 m).	4872 (Jan. 2016)		5224	
Activated carbon column made with FRP (total volume 6 m^3, packing bed volume 5.7 m^3, diameter 0.9 m).	11,339 (Jan. 2016)		12,157	
Nitrification reactor tank of 8.3 m^3 made with FRP with a stirring system.	11,509 (Jan. 2016)		12,339	
Centrifugal pump of 4 kW and 3600 rpm at 60 Hz. Height: 292 mm. Diameter: 265 mm	900 (Jan. 2016)		965	
Air diffuser	900 (Jan. 2016)		965	
Blower	5000 (Jan. 2016)		5361	
Air Jet-Venturi	900 (Jan. 2016)		965	AIR-VAC Engineering Company (2017)
Biogas analyser (H_2S, CH_4, O_2, CO_2)	14,700 (Jul. 2015)	650.4	15,760	MRU Instruments, Inc. (2015)

[a] CEPCI. Index for equipment. Value for July 2017: 686.7.

Table 5
Total fixed-capital investment for desulfurizing scenarios.

		% of FCI	Sc1.0	Sc2.0	Sc2.1//Sc2.2	Sc3.0	Sc4.0
Direct Costs	Purchased equipment cost (PEC)	30	35,745 €	52,480 €	33,815 €	21,949 €	28,882 €
	Installation	14	16,681 €	24,491 €	15,780 €	10,243 €	13,478 €
	Instrumentation and controls (installed)	8	9532 €	13,995 €	9017 €	5853 €	7702 €
	Piping (installed)	6	7149 €	10,496 €	6763 €	4390 €	5776 €
	Electrical systems (installed)	8	9532 €	13,995 €	9017 €	5853 €	7702 €
	Buildings	0	- €	- €	- €	- €	- €
	Yard improvements	0	- €	- €	- €	- €	- €
	Land	0	- €	- €	- €	- €	- €
Total direct costs		–	78,638 €	115,455 €	74,393 €	48,288 €	63,541 €
Indirect Costs	Engineering and supervision	13	15,489 €	22,741 €	14,653 €	9511 €	12,516 €
	Construction expense	8	9532 €	13,995 €	9017 €	5853 €	7702 €
	Contractor's fee	3	3574 €	5248 €	3381 €	2195 €	2888 €
	Contingence	10	11,915 €	17,493 €	11,272 €	7316 €	9627 €
Total Indirect costs		–	40,511 €	59,477 €	38,324 €	24,876 €	32,733 €
Total FCI		–	119,149 €	174,932 €	112,716 €	73,164 €	96,275 €

Table 6
Utility costs for the desulfurization plants operation.

Inputs and outputs				
Description	Cost (€ Units-1)	% Change CPI[a]	Costs (July 2017) (€ Units-1)	Source
Polypropylene Pall Ring (m^3)	637 (Jan. 2016)	2.8	654.87	Bioconservacion S. A. (2016)
Activated Carbon Non-regenerative (kg)	2.5 (Jan. 2016)	2.8	2.6	
NaOH (kmol)	72.0 (Jan. 2017)	−0.1	71.9	Haupold S.L. (2017)
NaHCO3 (kmol)	141.1 (Jan 2017)	−0.1	141.0	
H2O2 (kmol)	361.9 (Jan. 2017)	−0.1	361.5	Vadequímica S. L. (2017)
KOH (kmol)	130.0 (Feb. 2013)	1.2	131.6	(Lombardi and Carnevale, 2013)
NaNO3 (kmol)	38.3 (Sep. 2009)	9.9	42.0	BASF Española S.L (2009)
Electricity (MWh)	103.0 (Jan. 2017)	−0.1	102.9	(Eurostat, 2017)
Heat (MWh)	32.0 (Jan. 2017)	−0.1	32.0	
Industry cost labor of per year (25% of total salary)	–	–	9317.46	(INE, 2017)
Waste final disposal (tonnes)	23.0 (Jul. 2014)	1.0	23.2	(Moncada et al., 2018)
Maintenance costs (year)	1.5% of total direct cost			(Aguilera and Gutiérrez-Ortiz, 2016)

[a] Reported by INE (2017).

Sc2.2 was the workforce and chemicals consumption (77.3%). For the physical-chemical technologies in the Sc3.0 the chemical consumption accounted for 95.3% of the total cost of operation and for the Sc4.0 was mainly due to the activated carbon consumption (76.8%) and workforce (19.8%) (Table S3).

4. Conclusions

The environmental assessment of four desulfurization technologies shows that the most important stage is the daily desulfurization operation. Thus, the impact of construction and disposal was negligible. The best scenario was the aerobic BTF, although Sc2.0 (anoxic BTF) had slightly higher impact due to the construction of the nitrification bioreactor. Therefore, anoxic desulfurization could be an attractive alternative if the nitrification bioreactor is already present at the STP as this gives the lowest CO_2 emissions of 7.2 kg CO_2 kg^{-1} S-H_2S treated vs 13.9 kg CO_2 kg^{-1} S-H_2S treated for aerobic desulfurization. The chemical scrubber was

Table 7
OPEX costs of the different desulfurization technologies, Units: Euros per 1 kg of S-H_2S treated.

Scenario	Operational costs per 1 kg of S-H_2S treated (€)	Operational costs per m^3 of biogas treated (c€)
Sc1.0	5.20	2.04
Sc2.0	6.29	2.47
Sc2.1	4.35	1.71
Sc2.2	6.31	2.48
Sc3.0	84.92	33.39
Sc4.0	13.65	5.37

the worst desulfurization alternative because the emissions were higher – except for "Climate Change", which was slightly higher on using impregnated active carbon. The main inducers for the "Climate Change" were: electricity consumption in Sc1.0 and Sc2.0, H_2O_2 production in Sc3.0 and the carbon production and impregnation process in Sc4.0. Moreover, in all cases the contribution of sulfur compounds was less than 5%. The lowest operational costs were 2.04 and 1.71 c€ per m^3 of biogas for an aerobic BTF and anoxic BTF in Sc2.1.

Up to 230 or 411 kg CO_2 kg^{-1} S-H_2S could be saved with electricity and electricity and heat recovery, respectively, on using an aerobic BTF. In contrast, the direct combustion of biogas in a flare will emit 26 kg CO_2 kg^{-1} S-H_2S treated.

The physical-chemical technologies are mature processes and therefore it is quite difficult to reduce their high environmental impact, which is mainly due to the use of chemical compounds (chemical scrubber unit) and packing material (activated carbon unit). Physical-chemical technologies are used instead of biological processes because the latter are still not well known by biogas producers. In any case, a fast implementation is expected because biological processes have lower operational costs and they are more environmentally friendly than physical-chemical processes.

The results obtained in this study answer the question 'which biological technique is the best for biogas energy recovery in a CHP engine?' It was demonstrated that aerobic BTF had a lower impact and operational cost than anoxic BTF if it is necessary to build a nitrification bioreactor. However, if the nitrification bioreactor is already in place at the STP then anoxic BTF is the best approach.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jclepro.2018.02.018.

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